nvironmental scientists monitor fuel oil contamination of water and soil, ✓but indoor exposure has only recently come under investigation as a possible human health threat. Fuel oil is a complex mixture containing hundreds of hydrocarbon compounds such as benzene, toluene, and xylene. Inhaling or contacting fuel oil components can cause a variety of health problems, such as intoxication, headaches, hypertension, drowsiness, skin irritation, and disorders of the immune and reproductive systems. Such components occur in fuel oil in low concentrations of 0.5-5%, depending on the manufacturer. "But when you spill two hundred gallons

in a basement, that's a lot of toxic compounds," says David Tilotta, a chemistry professor at the University of North Dakota in Grand Forks.

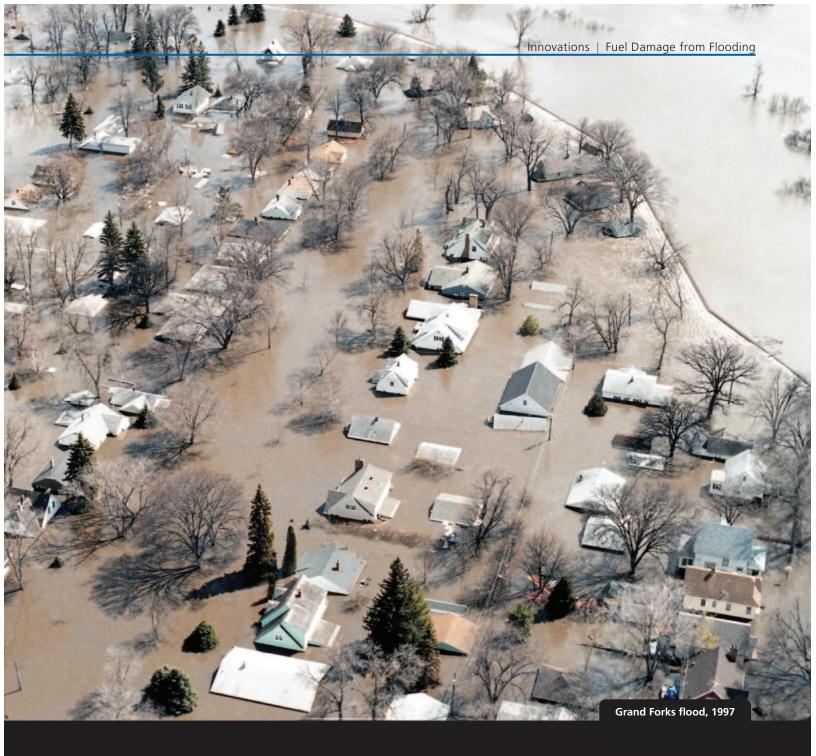
Tilotta set out to study indoor fuel oil contamination firsthand when the Red River flooded Grand Forks in April 1997. After documenting the problem, Tilotta teamed up with microbiologist Evguenii Kozliak, also a professor at the University of North Dakota, to probe why repeated cleanup procedures do not remove fuel oil from building materials such as concrete and wood, and what other cleanup possibilities there might be. What they've found may someday result in a commercial product that could

remove hydrocarbons and other stubborn hazardous compounds such as chemical warfare agents, explosives, and insecticides from solid materials.

### Overflowing with Curiosity

When the Red River flooded Grand Forks, 11,000 homes and businesses were swamped, and 50,000 of the town's 71,000 residents were evacuated from their homes. After the torrential waters subsided and the cleanup began, Tilotta noticed large amounts of smelly fuel oil coating homes, fences, soil, and other structures.

One large trail of fuel oil was traced back to a school that had filled its 3,000-gallon



outdoor tank just before the flood struck. In other cases, basements were contaminated when fuel oil leaked from indoor storage tanks (which keep fuel from solidifying into a gel form during the city's cold winters) or when fuel lines ruptured. According to records kept by the Grand Forks fire department, 1,200 homes reported problems with fuel oil spills ranging from 50 to 260 gallons.

None of the residents of Grand Forks reported that their postflood health symptoms were caused by fuel oil exposure. However, this does not surprise Tilotta, because after a flood, stress and anxiety levels run high, and people often blame symptoms on these culprits. Plus, other exposures

were occurring at the same time—many people clean with bleach to kill mold, and local hospitals reported health problems from people inhaling bleach fumes. "It's hard to separate whether a rise in blood pressure, a headache, or skin irritation is due to stress or inhaling bleach or fuel oil [fumes]," says Tilotta.

Many homeowners also try to mask flood-related odors with perfumed air fresheners, which may contribute to health problems similar to those caused by fuel oil vapors. Moreover, "like other odors, after a while you become desensitized to fuel oil and stop smelling it," says Wally Helland, environmental health supervisor at the Grand Forks Public Health Department. However, the effects don't disappear with the smell.

Tilotta, his curiosity piqued by the sight of the fuel-coated structures and neighbors' reports of lingering fuel oil odors, set out to investigate. He and Kozliak discovered that fuel oil hydrocarbons become trapped deep inside structural materials. The pressure of water is very high during a flood—the Red's flow rate during the flood reached 140,000 cubic feet per second, compared to the normal flow rate of 780 cubic feet per second—and it pushes hydrocarbons deep into the microscopic pores of wood and concrete.

The researchers quantified this effect by simply weighing pieces of concrete before and after soaking them in water; in concrete, 8–10% of the volume is accounted for by pores. Then they applied varied amounts of hydrocarbons to 1-gram pieces of concrete. They found that if they applied less than 2–2.5% hydrocarbon by volume, it was absorbed into the concrete rapidly and irreversibly.

Tilotta and Kozliak found that pore volume is even greater in wood. For wood, they let hydrocarbons applied on the surface diffuse through the piece of wood. Then they removed 5-centimeter chunks of the wood and analyzed them for the hydro-

carbon. Within a matter of hours, hydrocarbons had penetrated a few centimeters into the wood.

Consequently, even rigorous surface cleanings with pressure washers, bleach, or other methods cannot penetrate enough to eliminate the fuel oil contaminants. "If it soaks into floor joists or other porous material, fuel oil cannot be removed with any cleaning method," says Helland. In some Grand Forks homes, the damage was limited to wooden structures such as basement staircases, which were removed and replaced. But in others, the contamination was so extensive that the buildings had to be demolished.

### An Inside Job

Tilotta continued his inquiry by measuring concentrations of airborne fuel oil components in three homes about nine months after the flood, during the winter, when "the houses were shut up and fumes were distributed everywhere," he says. These homes were

selected because the owners reported still smelling fuel oil, even though they had scoured the buildings with pressure washers, bleach, detergents, and other common cleaning methods.

Tilotta used the best available technology at the time—portable chromatography monitors that were set up in a home and run overnight, much like a professional radon test. According to Tilotta's evaluation, volatile fuel oil components were still detectable. However, the portable monitors were intended to measure volatile hydrocarbons in gasoline, not the heavier hydrocarbons in fuel oil. So, although the values obtained showed that petroleum hydrocarbons were qualitatively present, Tilotta believes the quantitative readings were inaccurate. (Because of this need for

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better validation of fuel oil components in indoor air, Tilotta later received a grant from the U.S. Environmental Protection Agency [EPA] to find ways to better quantitate hydrocarbons specific to fuel oil.)

Tilotta's preliminary study prompted the Grand Forks Public Health Department to request help from the EPA in investigating further, in more homes, and in helping to determine some kind of limits to advise people about safety and health problems. "I had never heard of indoor fuel oil contamination, and even the EPA didn't have any good background on it," says Helland.

Experts from the EPA conducted a study of 34 homes about one year after the flood occurred. Six homes still had measurable hydrocarbon vapors that were considered a serious health problem; the

cause structural damage to housing and heating systems, releasing fuel oil into flood waters (above). In flooded basements, oilcontaminated water can seep into walls and stairwells, later releasing toxic hydrocarbons into indoor air (below).

Oil and water don't mix. Flooding can

homeowners were advised to move or undergo major structural work to replace contaminated structures.

Like most states, North Dakota did not have a safety limit for indoor inhalational exposure to volatile hydrocarbons in fuel oil before the devastating flood. Based on numbers in the medical literature and looking at what a few other states had done, health experts in North Dakota in 1998 set a limit of 2.4 milligrams per cubic meter of total fuel oil in air for sensitive populations—pregnant women, the elderly, and children—and 5 milligrams per cubic meter for the general population.

### **Eating Away at Contamination**

Because Tilotta's laboratory tracks environmental pollutants in water and soil, he knew that certain bacteria serve as bioremediation agents. He drew on Kozliak's expertise to explore the use of microbes to clean up lingering fuel oil contamination in concrete and wood. Researchers in Kozliak's laboratory isolated strains of the common and harmless soil and water bacterium *Pseudomonas aeruginosa*, grew them in the laboratory, and then applied the microbes to contaminated samples of wood and concrete.

In the first experiment, building-grade concrete samples were saturated with naphthalene or n-hexadecane, two representative hydrocarbons found in fuel oil. The chemicals were radiolabeled with carbon-14 (<sup>14</sup>C) to monitor their movement out of the concrete. From a local soil site contaminated with waste railroad engine oil, the researchers obtained strains of *P. aeruginosa* that consume either naphthalene or n-hexadecane, biodegrading the chemicals into measurable metabolites such as carbon dioxide. *P. aeruginosa* was applied to the samples in three forms—as an aqueous solution, on filter paper, or on agar (a gel).

The bacteria soon went to work "eating" the radiolabeled contaminants. Kozliak explains that by feeding on hydrocarbons on the surface, bacteria create a gradient that allows hydrocarbons deep inside pores to migrate to the surface more quickly than they would naturally. This helps to accelerate the diffusion of hydrocarbons trapped inside. Once they make it to the surface, these hydrocarbons either are degraded by the bacteria or evaporate, diffusing into the air at safe concentrations.

The aqueous solution of bacteria proved the poorest way to eliminate hydrocarbons from the concrete chips. After seven days of incubation, just 19% of the n-hexadecane and 35% of the naphthalene was removed. Other researchers have reported that water hinders the diffusion of volatile organic compounds in concrete and soil, which parallels Kozliak's finding that aqueous solutions of bacteria inefficiently degraded hydrocarbons. "When pores fill with water, volatile organic compounds cannot volatilize and escape," explains Kozliak.

In contrast, applying bacteria on filter paper or agar removed 80% of n-hexadecane and 55% of naphthalene. The results of this study will be published in a future issue of the journal *Acta Biotechnologica*.

In the second experiment, pieces of Southern yellow pine, a common wood for home construction, were soaked with <sup>14</sup>C-labeled naphthalene. Strains of *P. aeruginosa* that consume naphthalene were applied either as an aqueous solution, on filter paper, or on agar. Once again, the

filter paper and agar proved superior to solutions of bacteria. Between 90% and 98% of the naphthalene was removed in two days with either filter paper or agar treatments, compared to just 82% removed by a solution of bacteria after seven days of treatment. This information was presented at the Sixth International Symposium for Environmental Biotechnology, held in Veracruz, Mexico, in June 2002, and is being prepared for journal publication.

The researchers also saturated a piece of wood with fuel oil (rather than just two of its components), treated it with *P. aeruginosa*, and monitored the changes in 200 hydrocarbon components of the fuel oil by chromatographic techniques. All 200 compounds were reduced to varying degrees over a three-week treatment period, says Kozliak. So far, he's analyzed only straightchain hydrocarbons with 10–20 carbons (such as dodecane). These compounds were reduced by an average of 75%.

These preliminary proof-of-concept experiments are the first step toward a commercial product that could rid solid materials of hydrocarbons and other hazardous compounds including chemical warfare agents, explosives, and insecticides. Kozliak foresees bacteria being applied to contaminated surfaces in a gel or pellet form, methods that mimic the laboratory application by agar or filter paper. Patent applications on the technology have been filed by the University of North Dakota.

This research on contaminated concrete and wood is "very relevant to the conditions in regions with floods," says Sergio Revah, a chemical engineer who works with microbial degradation of recalcitrant compounds at Universidad Autonoma Metropolitana in Iztapalapa, Mexico. Although many details remain to be worked out, such as the best types of microorganisms and how to apply them, the approach "may lead to interesting bioremediation technologies," Revah says.

Carol Potera

## Suggested Reading

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